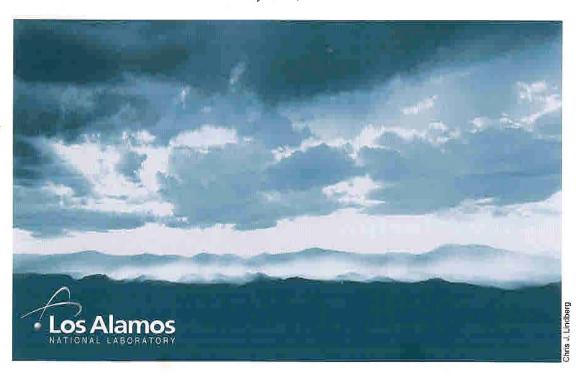
# **Utility of Monte Carlo Modelling for Holdup Measurements**

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## Utility of Monte Carlo Modelling for Holdup Measurements

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#### **ABSTRACT**

Non-destructive assay (NDA) measurements performed to locate and quantify holdup in the Oak Ridge K25 enrichment cascade used neutron totals counting and low-resolution gamma-ray spectroscopy. This facility housed the gaseous diffusion process for enrichment of uranium, in the form of UF<sub>6</sub> gas, from ~20% to 93%. Inventory of <sup>235</sup>U inventory in K-25 is all holdup. These buildings have been slated for decontamination and decommissioning. The NDA measurements establish the inventory quantities and will be used to assure criticality safety and meet criteria for waste analysis and transportation. The tendency to err on the side of conservatism for the sake of criticality safety in specifying total NDA uncertainty argues, in the interests of safety and costs, for obtaining the best possible value of uncertainty at the conservative confidence level for each item of process equipment. Variable deposit distribution is a complex systematic effect (i.e., determined by multiple independent variables) on the portable NDA results for very large and bulky converters that contributes greatly to total uncertainty for holdup in converters measured by gamma or neutron NDA methods. Because the magnitudes of complex systematic effects are difficult to estimate, computational tools are important for evaluating those that are large. Motivated by very large discrepancies between gamma and neutron measurements of high-mass converters with gamma results tending to dominate, the Monte Carlo code MCNP has been used to determine the systematic effects of deposit distribution on gamma and neutron results for <sup>235</sup>U holdup mass in converters. This paper details the numerical methodology used to evaluate large systematic effects unique to each measurement type, validates the methodology by comparison with measurements, and discusses how modelling tools can supplement the calibration of instruments used for holdup measurements by providing realistic values at well-defined confidence levels for dominating systematic effects.

#### INTRODUCTION

An in-depth guide to holdup measurements using portable gamma-ray measurement equipment is given in reference [1]. In general, holdup measurements are difficult mainly because of the infinite possibilities for deposit geometry. This difficulty is somewhat minimized by taking a generalized approach in which every deposit is modelled as a point, line, or area. This Generalized Geometry Holdup method (GGH) is used extensively in the DOE complex for measuring holdup with portable gamma-ray equipment, and requires the user to setup the measurement such that, within the field-of-view of the detector, the deposit resembles one of the three geometries.

Determining the realistic systematic effects of many individual parameters that influence the analysis result of portable holdup measurements is straightforward for many types of equipment. Examples of some of those parameters are: <sup>235</sup>U enrichment, mass attenuation coefficient of the

deposit, deposit width, equipment (container) thickness, equipment length, etc. However, the systematic effects of departure from the assumption of uniform deposit thickness can be difficult to quantify in some cases. Because a non-uniform deposit distribution in an extended duct measured at only one position rather than 100 departs in a non-quantifiable way from the calibration assumption of uniformity, the magnitude of the systematic effect in a realistic range of non-uniformity must be determined as part of the systematic error in the measurement. This is complex because two effects dominate. One is the non-uniformity in the field of view of the single measurement. The second is the non-uniformity over the full length of the duct. The complexity can be reduced by treating the two as separate and independent systematic effects using realistic assumptions about the range of deposit thickness in the particular vacuum line. It is not always possible to reduce a complex systematic effect of deposit distribution to multiple simple effects. The use of numerical techniques such as Monte Carlo can determine the complex effects in such cases. An example is taken from measurements of converters used to enrich uranium as UF<sub>6</sub> in a gaseous diffusion process.

#### K-25 GASEOUS DIFFUSION PLANT

The K25/K27 uranium gaseous diffusion plant at Oak Ridge National Laboratory is currently scheduled for decontamination and decommissioning (D&D). In the late 1980's, an extensive project was undertaken to quantify the mass and location of uranium holdup deposits within the process equipment using non-destructive assay (NDA) techniques. Both neutron and gamma-ray (at 186 keV) measurements were made on all process piping and equipment. The authors of this paper were asked to conduct an assessment [2] of this NDA measurement campaign and advise on its usefulness for supporting D&D activities.

The focus of this paper is the results of the NDA measurements on converters. Converters are large cylindrical pieces of equipment with several hundred small tubes inside through which gaseous UF<sub>6</sub> is passed. Figure 1 [3] shows a typical converter.

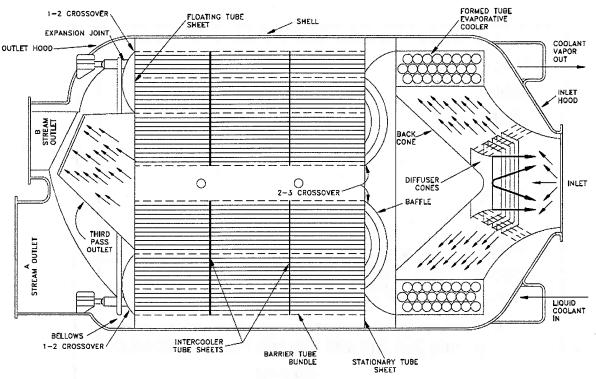


Figure 1. Cut-away view of a typical gaseous diffusion converter.

Their complex design and extended dimensions add significant uncertainty to portable NDA measurements including those that use neutrons but especially those based on gammas. The tendency for deposit models and assumptions to underestimate gamma attenuation combined with numerous effects (including moderation, scattering, and alternative chemical forms) that tend to enhance neutron signals relative to the calibrated response would tend to shift the balance to high neutron results relative to gamma for the same converter. Just the opposite was observed in the results for the so called "high-mass" converters (> 200 g). Figure 2 is a plot of the neutron mass results vs the gamma-ray mass results. As seen in the plot most of the gamma-ray NDA masses are greater than the neutron NDA masses.

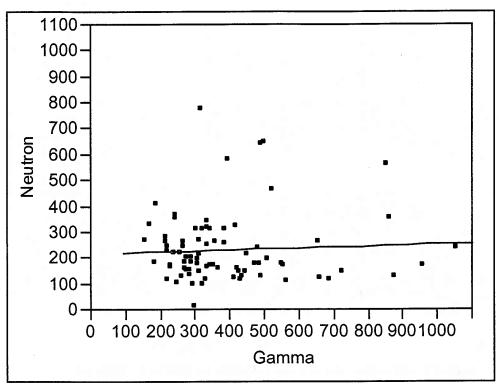


Figure 2

Thus, the existence of a very large systematic effect other than the previous typical examples is indicated. The absence of any visible or analytically detectable correlation between neutron and gamma results, suggests that the effect is large, and possibly complex (influenced by more than one independent parameter) in that the expected correlation between the two measurements is washed out. A variable effect (sometimes shifting gamma results low relative to neutron) can also explain the occasional high neutron result. Deposit distribution effects can be large, complex and variable.

A distribution of uranium in the converter that differs from the calibration assumption is a plausible systematic effect that could lead to large gamma-neutron discrepancy. This seems particularly apparent for the larger Type 1 and 2 converters because the gamma and neutron detectors are positioned differently in the horizontal dimension as illustrated in Figure 3. The calibration was developed for horizontally uniform deposits. If deposits concentrate toward the horizontal centre of the converter, neutron results for the same deposit will be higher than gamma results. If deposits concentrate toward either or both horizontal ends of the converter, as illustrated by the red curve in Figure 3, gamma results for the same deposit will be higher than neutron results. Subsequent measurements on a few converters, taking data at the original four measurement locations and at the top and bottom in the middle of the converter, showed no evidence of a non-uniform deposit in the axial direction.

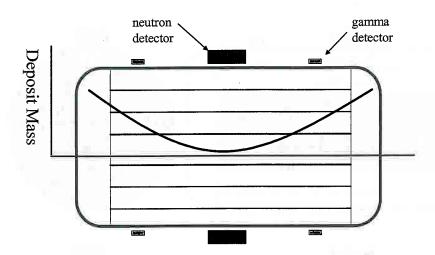


Figure 3

#### MCNP SIMULATION OF DEPOSIT GEOMETRY

The possibility of a non-uniform deposit in the radial dimension was considered next. The calibration for the NDA equipment was actually developed for a radially non-uniform deposit residing in the first-pass region of the converter only (Figure 4a). This deposit model was selected because the corresponding calibration is conservative, giving a higher mass result than the true mass if a measured deposit tends to be uniformly distributed throughout the (first-, second- and third-pass) volumes of the converter, for example. Figure 4b illustrates the uniform-deposit model. In the absence of attenuation, the effect on gamma and neutron measurements of rearranging a constant mass of <sup>235</sup>U between these two (first-pass and uniform) deposit distributions might be very similar for the two types of measurements. However, internal converter hardware attenuates the 186-keV gamma rays significantly so that redistributing deposits from first-pass to uniform enhances the gamma result more than the neutron because of reduced gamma attenuation.

It is very difficult to test the radial deposit distribution hypothesis empirically. Therefore, three distributions of the same <sup>235</sup>U deposit mass were modelled along with the gamma and neutron detectors and the Type-2 converter. Monte Carlo techniques were used to determine the gamma and neutron responses to each deposit distribution. Two of the three distributions were the first-pass and uniform distributions, and the third was the shell distribution, where all the material was deposited on the inner shell of the converter, shown in Figure 4c. The results of the Monte Carlo simulations for the gamma and neutron responses are given in Table 1 as ratios to the first-pass response because it represents the calibration model for these detectors.

The results in Table 1 reveal that the gamma results are enormously enhanced as the deposit distribution shifts toward uniform from the first-pass calibration model while the corresponding enhancement in the neutron result is small. If a uniform deposit in the Type-2 converter is measured, the gamma result would be biased by +163% while the neutron bias would be only +16% for the uniform deposit distribution. Should the deposit distribution shift from uniform toward the containment shell of the converter, the bias for the neutron result remains less than +37% while the gamma bias approaches +500%.

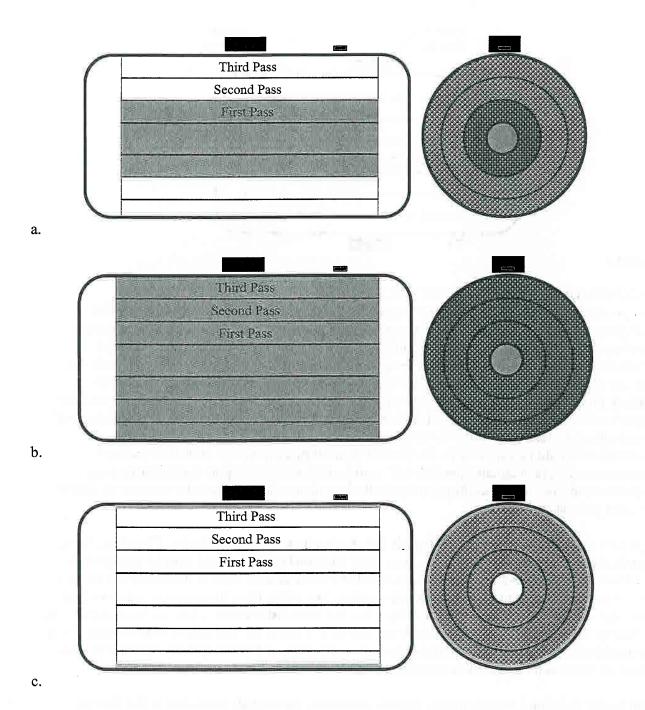


Figure 4. Three sketches of the longitudinal (left) and transverse (right) cross sections of the Type-2 converter show the a) first pass, b) uniform, and c) shell models for radial distribution of uranium within the converter. Red shading indicates the uranium deposit. Refer to Figure 3 for information and a key on the gamma and neutron detectors and their positions.

**Table 1.** Monte Carlo Simulated Gamma and Neutron Response for Type-2 Converter: Three Deposit Distribution Models

Response Ratio (to First-pass Response)	Neutron	% NDA Bias	Gamma	% NDA Bias
Uniform/First Pass	1.16	16%	2.63	163%
Shell/First Pass	1.37	37%	5.87	487%

This Monte Carlo result reinforces the likelihood that the uniform model for deposit distribution is better suited to actual deposits than the first-pass model because the model illustrates that deposits of <sup>235</sup>U more-or-less uniformly distributed throughout the volume of the Type-2 converter can account for the observed gamma-neutron discrepancies where the gamma result exceeds that for neutrons. A "tighter axial" distribution than that shown in Figure 4a could explain the occasional result in which neutron exceeds gamma.

The original measured and model adjusted masses of 12 converters are listed in Table2 for both gamma-ray and neutron measurements. The ratios of the gamma to neutron measured masses are shown in Table 3 for each model.

Table 2. Converter masses for both gamma-ray and neutron measurements

Gamma-ray Results			Neutron Results		
	Mass Adjusted for	Mass Adjusted for		Mass Adjusted for	Mass Adjusted for
Mass (g)	Uniform Deposit (g)	Shell Deposit (g)	Mass (g)	Uniform Deposit (g)	Shell Deposit (g)
3725	1416	635	771	665	563
3499	1330	596	417	359	304
1056	402	180	233	201	170
959	365	163	171	147	125
877	333	149	126	109	92
864	329	147	352	303	257
853	324	145	558	481	407
728	277	124	143	123	104
690	262	118	. 114	98	83
663	252	113	119	103	87
657	250	112	260	224	190
571	217	97	1950	1681	1423

Table 3. Ratios of gamma-ray and neutron masses for each deposit model

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	First Pass	Uniform	Shell
	4.83	2.13	1.13
	8.39	3.70	1.96
	4.53	2.00	1.06
Ì	5.61	2.47	1.31
	6.96	3.07	1.62
	2.45	1.08	0.57
	1.53	0.67	0.36
	5.09	2.25	1.19
١	6.05	2.67	1.41
١	5.57	2.46	1.30
١	2.53	1.11	0.59
١	0.29	0.13	0.07
•			•

As seen in Table 3, the alternate deposit models result in gamma-to-neutron mass ratios that are closer to unity than the first pass model.

### MODELLING OF PROCESS EQUPMENT

Models of complex process equipment could be developed to verify and improve the calibration of NDA measurement equipment used for holdup. This could greatly improve the quality of holdup measurements. In addition, with access to uncontaminated process equipment and sealed sources that are representative of the facility process material the equipment models could be further improved.

## **CONCLUSIONS**

Monte Carlo modelling can greatly improve the accuracy of holdup measurements of complex process equipment.